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*Supplement of*

# A spatiotemporally separated framework for reconstructing the sources of atmospheric radionuclide releases

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## Note S1: Bayesian method for source reconstruction

The Bayesian method is combined with Markov chain Monte Carlo sampling to estimate the source location and release rate simultaneously. Bayes' theorem can be expressed as:

$$p(\boldsymbol{s}|\boldsymbol{\mu}) \propto p(\boldsymbol{\mu}|\boldsymbol{s})p(\boldsymbol{s}) , \tag{S1}$$

where $\boldsymbol{s}$ is the parameter vector containing source parameters and $\boldsymbol{\mu}$ is an observation vector. $p(\boldsymbol{s})$ describes the probability distribution of prior knowledge on $\boldsymbol{s}$ and $p(\boldsymbol{\mu}|\boldsymbol{s})$ is the likelihood function quantifying the goodness of fit between the simulations and observations. Consistent with general approaches, we define $p(\boldsymbol{s})$ as the uniform distribution bounded by the lower and upper limits of the source parameters. Referring to (Dumont Le Brazidec et al., 2021), $p(\boldsymbol{\mu}|\boldsymbol{s})$ is defined as the log-Cauchy distribution:

$$p(\boldsymbol{\mu}|\boldsymbol{s}) = -\sum_{i=1}^{O} ln\left\{r + \left[ln(\mu_i + \mu_t) - ln\left(\left(F(\boldsymbol{s})\right)_i + \mu_t\right)\right]^2\right\} + \frac{1}{2}ln(r) , \tag{S2}$$

where $O$ is the number of observations; $F$ is a function that maps the source parameters to the simulations; $r$ is a covariance parameter which forms the covariance matrix $\mathbf{R} = r\mathbf{I}$ and $\mu_t$ is a positive threshold that ensures the logarithm is defined properly for zero measurements or simulations.

However, the release rate is time-varied, so it is not realistic to define the prior distribution of the release rate in every time step. Hence, we incorporate the source inversion method into this process, which involves calculating the corresponding release rate for the sampled source location and then obtaining Eq. (S3) using the sampled location and calculated release rate. We apply a traditional Tikhonov method to calculate the release rate as follows:

$$\boldsymbol{q}(\boldsymbol{r}) = argmin\left(\frac{1}{2}\|\boldsymbol{\mu} - \mathbf{A}(\boldsymbol{r})\boldsymbol{q}\|_2^2 + \frac{1}{2}\lambda^2\|\boldsymbol{q}\|_2^2\right) , \tag{S3}$$

where $\boldsymbol{q}(\boldsymbol{r})$ refers to the estimated release rate vector under the source location $\boldsymbol{r}$ and $\mathbf{A}(\boldsymbol{r})$ refers to the source–receptor matrix given $\boldsymbol{r}$. $\lambda$ is a regularization parameter that is automatically selected by generalized cross-validation (Hansen and O'Leary, 1993).

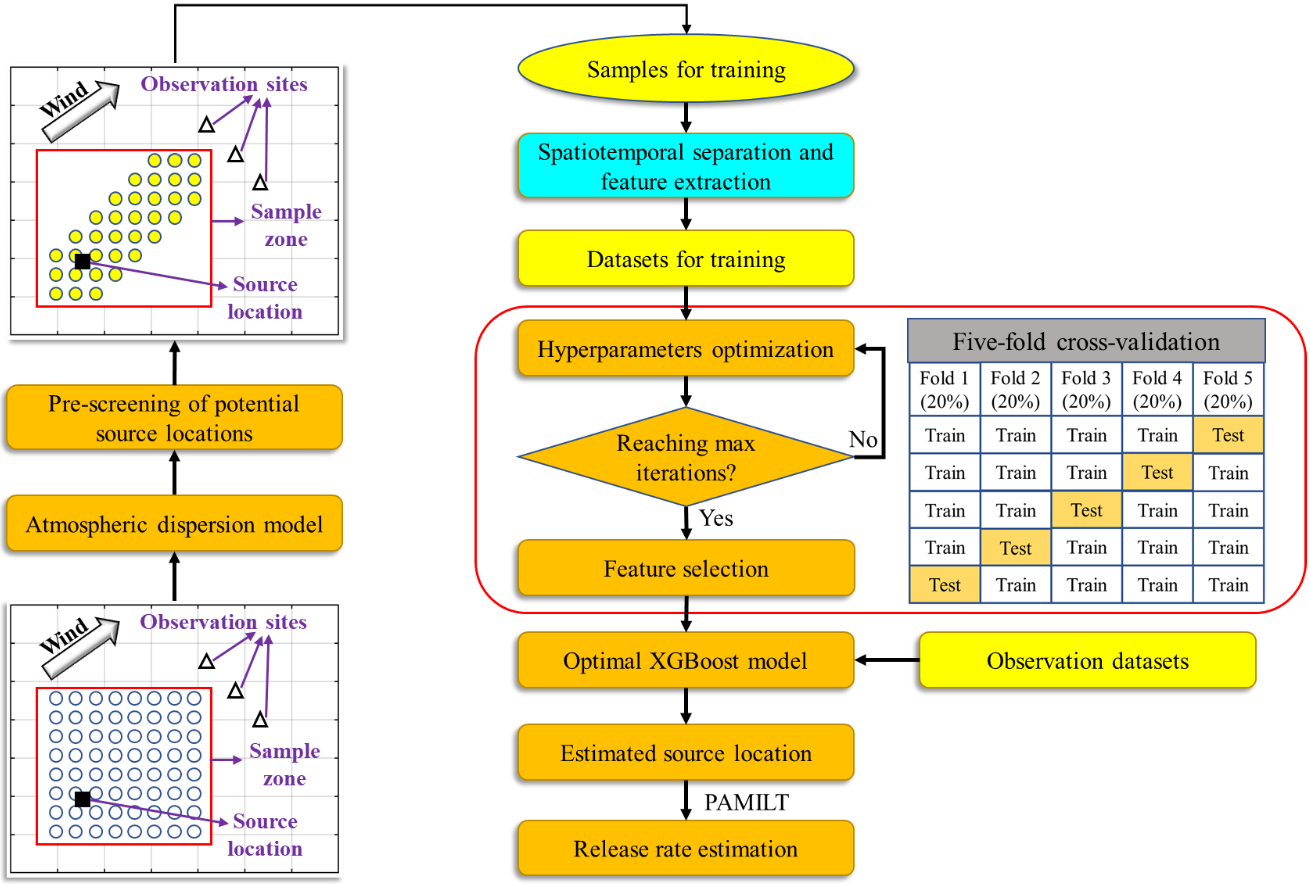


**Figure S1.** Flowchart of the proposed spatiotemporally separated source reconstruction method.

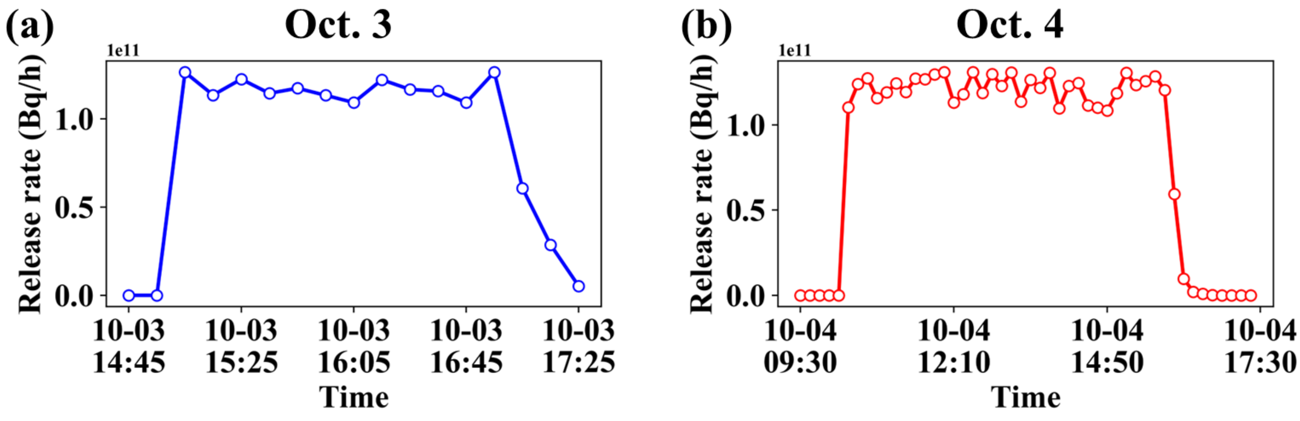


**Figure S2.** Synthetic release rates for generating synthetic observations of SCK CEN $^{41}Ar$ experiment. (a) 3 October; (b) 4 October.

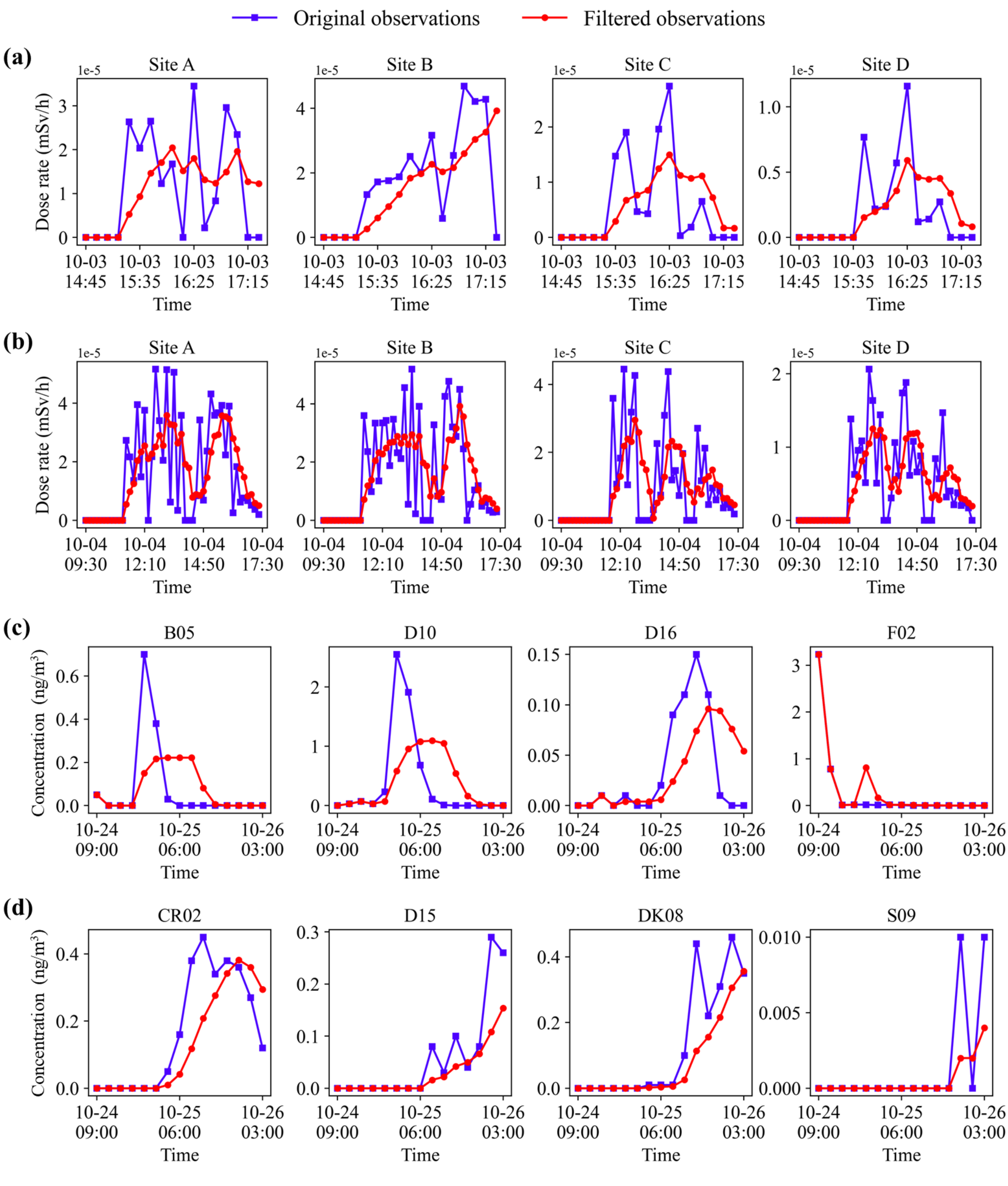


**Figure S3.** Observations before and after filtering at observation sites. SCK CEN $^{41}Ar$ experiment: (a) 3 October; (b) 4 October. ETEX-1 experiment: (c) Group 1; (d) Group 2.

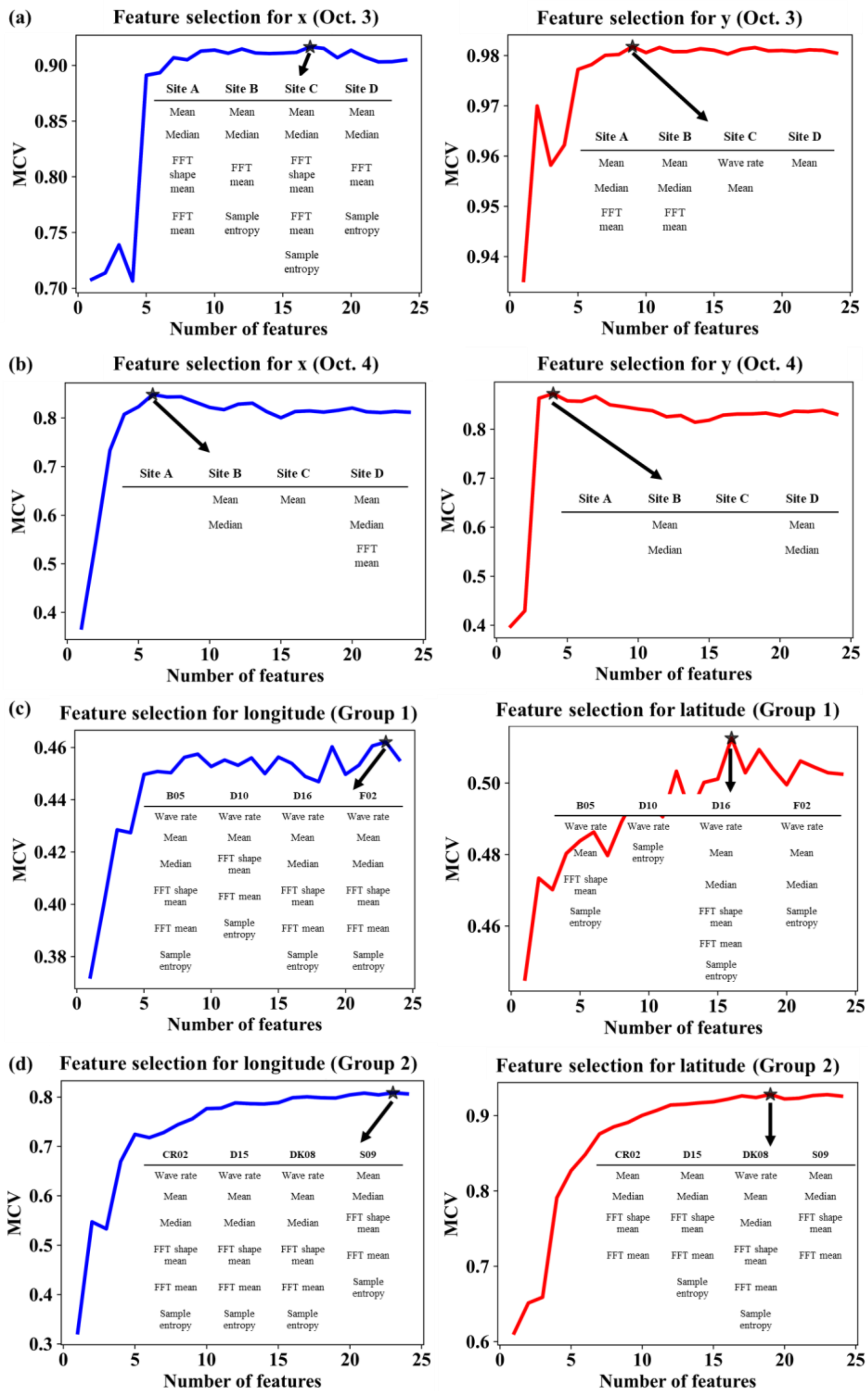


**Figure S4.** Results of feature selection in x (longitude) and y (latitude) directions. SCK CEN $^{41}Ar$ experiment: (a) 3 October; (b) 4 October. ETEX-1 experiment: (c) Group 1; (d) Group 2. The black stars denote the optimal number of features. The table inserted in each subgraph lists the selected features for each observation site.

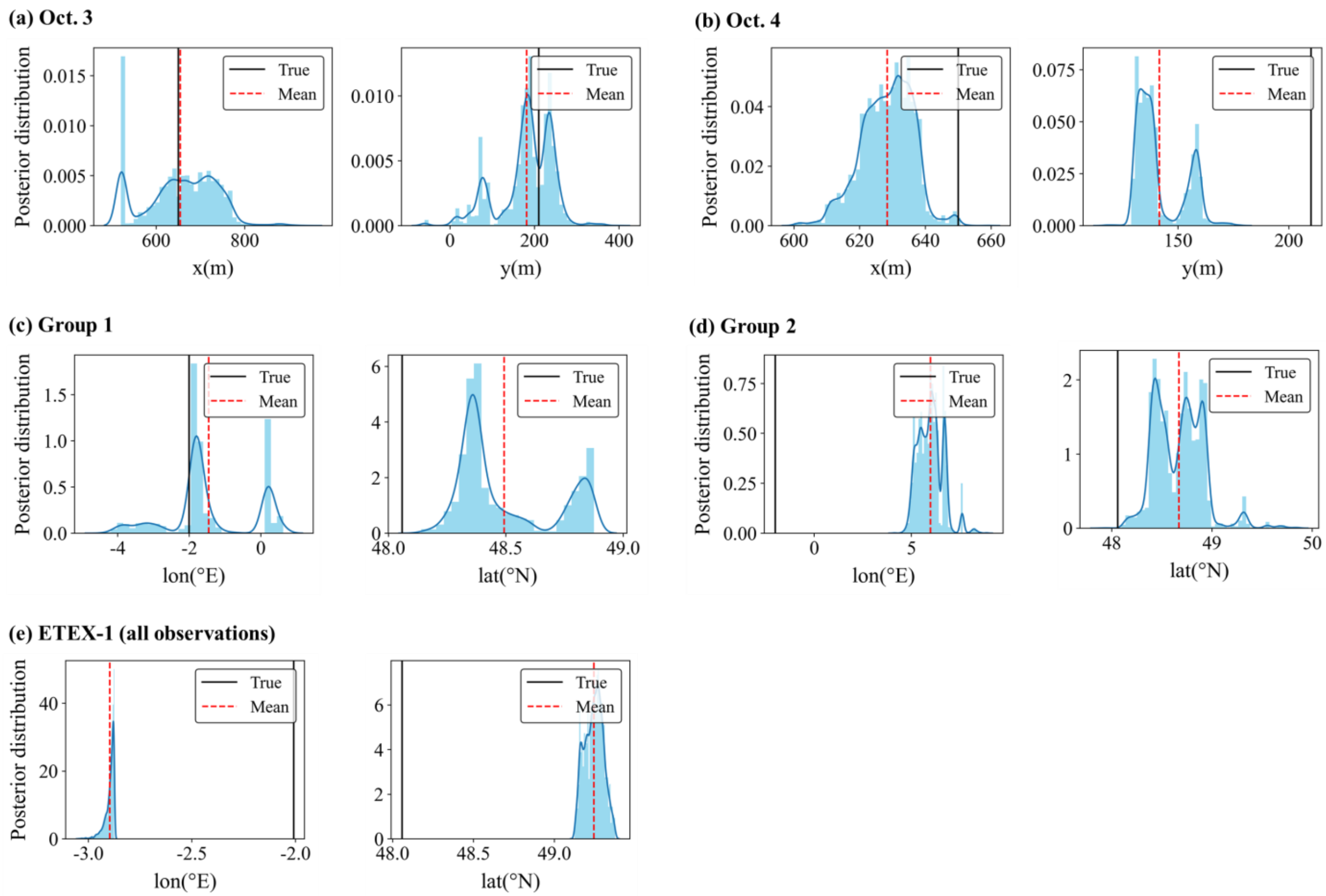


**Figure S5.** Posterior distributions of source location parameters. SCK CEN $^{41}Ar$ experiment: (a) 3 October; (b) 4 October. ETEX-1 experiment: (c) Group 1; (d) Group 2; (e) ETEX-1 (all observations in ETEX-1 are used). The black solid lines denote the true location parameters and the dashed lines denote the mean estimates of all posterior samples.

**Table S1.** Hyperparameter optimization results. *max_depth*-maximum depth of a decision tree; *learning_rate*-step size shrinkage when updating; *n_estimators*-number of decision trees; *min_child_weight*-minimum sum of sample weight of a child node; *subsample*-subsample ratio of the training samples; *colsample_bytree*-subsample ratio of columns when constructing a decision tree; *reg_lambda*-L2 regularization term on weights; and *gamma*-minimum loss reduction required to split the decision tree.

| Optimization results | | SCK CEN $^{41}Ar$ experiment | | ETEX-1 experiment | |
|---|---|---|---|---|---|
| | | 3 October | 4 October | Group 1 | Group 2 |
| Hyperparameters | *max_depth* ([3,8]) | 8 | 7 | 3 | 7 |
| | *learning_rate* ([0.05,0.3]) | 0.07057 | 0.14413 | 0.06860 | 0.07619 |
| | *n_estimators* ([50,300]) | 283 | 185 | 93 | 234 |
| | *min_child_weight* ([2,10]) | 4 | 10 | 8 | 6 |
| | *subsample* ([0.5,1]) | 0.62353 | 0.52721 | 0.66447 | 0.62146 |
| | *colsample_bytree* ([0.01,1]) | 0.39145 | 0.57415 | 0.33031 | 0.77954 |
| | *reg_lambda* ([0.01,5]) | 0.71074 | 2.30624 | 3.95098 | 4.62217 |
| | *gamma* ([0.01,1]) | 0.47779 | 0.51660 | 0.67626 | 0.85628 |
| Optimal GC | | 0.01230 | 0.03700 | 0.88040 | 0.44510 |

**Table S2.** Hyperparameter optimization results of all 50 runs for 3 October of SCK CEN $^{41}$Ar experiment.

| Run | Hyperparameters | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | *max_depth* | *learning_rate* | *n_estimators* | *min_child_weight* | *subsample* | *colsample_bytree* | *reg_lambda* | *gamma* |
| 1 | 8 | 0.06963 | 257 | 2 | 0.58442 | 0.99833 | 0.56084 | 0.03347 |
| 2 | 8 | 0.05003 | 261 | 3 | 0.65774 | 0.76821 | 3.67031 | 0.40337 |
| 3 | 7 | 0.08651 | 246 | 4 | 0.76497 | 0.86844 | 1.82068 | 0.02107 |
| 4 | 3 | 0.10114 | 240 | 3 | 0.72483 | 0.99964 | 1.90473 | 0.40321 |
| 5 | 4 | 0.09505 | 299 | 4 | 0.86627 | 0.91229 | 2.72513 | 0.73273 |
| 6 | 6 | 0.12811 | 198 | 4 | 0.86151 | 0.91167 | 2.01739 | 0.64585 |
| 7 | 8 | 0.12143 | 160 | 5 | 0.76193 | 0.87631 | 1.32947 | 0.10111 |
| 8 | 4 | 0.10118 | 149 | 2 | 0.75307 | 0.47997 | 1.42352 | 0.32648 |
| 9 | 6 | 0.08344 | 203 | 2 | 0.73900 | 0.58051 | 4.27579 | 0.36316 |
| 10 | 6 | 0.08371 | 293 | 4 | 0.70524 | 0.50305 | 1.58921 | 0.90349 |
| 11 | 6 | 0.08044 | 203 | 7 | 0.74233 | 0.83712 | 1.87067 | 0.66921 |
| 12 | 8 | 0.08750 | 298 | 5 | 0.73452 | 0.95439 | 3.24463 | 0.23793 |
| 13 | 8 | 0.12917 | 218 | 5 | 0.64402 | 0.57828 | 0.79801 | 0.43434 |
| 14 | 5 | 0.09389 | 279 | 2 | 0.97316 | 0.80680 | 1.61133 | 0.05062 |
| 15 | 8 | 0.14586 | 255 | 9 | 0.54883 | 0.74530 | 3.62691 | 0.21478 |
| 16 | 8 | 0.09194 | 160 | 3 | 0.59974 | 0.83406 | 0.33249 | 0.18032 |
| 17 | 4 | 0.08920 | 257 | 2 | 0.67346 | 0.99730 | 0.98970 | 0.17230 |
| 18 | 4 | 0.09419 | 294 | 4 | 0.79714 | 0.87812 | 3.77772 | 0.88406 |
| 19 | 5 | 0.07604 | 299 | 3 | 0.79858 | 0.83297 | 0.36589 | 0.27070 |
| 20 | 6 | 0.08451 | 231 | 3 | 0.76571 | 0.89974 | 2.67871 | 0.31997 |
| 21 | 5 | 0.15257 | 203 | 3 | 0.83687 | 0.94582 | 1.67365 | 0.06759 |
| 22 | 4 | 0.14711 | 180 | 3 | 0.82554 | 0.79287 | 1.10286 | 0.31295 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 23 | 5 | 0.08729 | 285 | 2 | 0.67684 | 0.91908 | 0.81695 | 0.76090 |
| 24 | 8 | 0.09440 | 235 | 2 | 0.66775 | 0.88929 | 4.40930 | 0.04806 |
| 25 | 7 | 0.10085 | 216 | 3 | 0.58725 | 0.68488 | 1.70407 | 0.25164 |
| 26 | 8 | 0.09937 | 200 | 3 | 0.83402 | 0.78555 | 3.59830 | 0.55999 |
| 27 | 3 | 0.12772 | 189 | 6 | 0.75408 | 0.99256 | 1.67164 | 0.24484 |
| 28 | 8 | 0.10973 | 183 | 2 | 0.50393 | 0.53818 | 0.67395 | 0.18678 |
| 29 | 6 | 0.09468 | 185 | 2 | 0.59535 | 0.75381 | 2.10634 | 0.48731 |
| 30 | 6 | 0.09652 | 247 | 3 | 0.69860 | 0.95369 | 0.05146 | 0.48637 |
| 31 | 7 | 0.06846 | 185 | 5 | 0.52549 | 0.61305 | 0.97320 | 0.17339 |
| 32 | 8 | 0.09323 | 215 | 4 | 0.74269 | 0.98432 | 4.30255 | 0.28215 |
| 33 | 7 | 0.09339 | 299 | 6 | 0.61681 | 0.49190 | 2.27687 | 0.96352 |
| 34 | 7 | 0.11693 | 234 | 3 | 0.74464 | 0.54387 | 1.02597 | 0.63504 |
| 35 | 4 | 0.06858 | 277 | 2 | 0.74264 | 0.92278 | 1.30424 | 0.81347 |
| 36 | 5 | 0.08068 | 246 | 5 | 0.73714 | 0.99006 | 1.39783 | 0.27963 |
| 37 | 8 | 0.08645 | 274 | 4 | 0.82352 | 0.99618 | 3.59875 | 0.82528 |
| 38 | 7 | 0.18011 | 226 | 8 | 0.66425 | 0.81094 | 0.98036 | 0.11274 |
| 39 | 5 | 0.08397 | 212 | 3 | 0.62934 | 0.45110 | 1.94896 | 0.64913 |
| 40 | 5 | 0.07800 | 228 | 2 | 0.66806 | 0.91700 | 0.32409 | 0.53206 |
| 41 | 6 | 0.07905 | 231 | 4 | 0.63064 | 0.93657 | 0.01082 | 0.03863 |
| 42 | 3 | 0.09277 | 261 | 3 | 0.72093 | 0.96486 | 1.73917 | 0.39009 |
| 43 | 5 | 0.08732 | 217 | 2 | 0.81405 | 0.78575 | 1.71376 | 0.85775 |
| 44 | 5 | 0.08417 | 225 | 3 | 0.61443 | 0.61703 | 2.06192 | 0.93001 |
| 45 | 8 | 0.14916 | 188 | 4 | 0.71686 | 0.87552 | 0.21908 | 0.58120 |
| 46 | 6 | 0.10745 | 179 | 3 | 0.82311 | 0.92434 | 3.99176 | 0.29124 |
| 47 | 4 | 0.13632 | 252 | 4 | 0.83077 | 0.92543 | 3.17264 | 0.31258 |
| 48 | 8 | 0.12402 | 176 | 4 | 0.70048 | 0.75866 | 3.18949 | 0.92647 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 49 | 8 | 0.07057 | 283 | 4 | 0.62353 | 0.39145 | 0.71074 | 0.47779 |
| 50 | 5 | 0.11104 | 197 | 3 | 0.79114 | 0.86436 | 3.16004 | 0.19049 |

**Table S3.** Hyperparameter optimization results of all 50 runs for 4 October of SCK CEN $^{41}Ar$ experiment.

| Run | Hyperparameters | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | *max_depth* | *learning_rate* | *n_estimators* | *min_child_weight* | *subsample* | *colsample_bytree* | *reg_lambda* | *gamma* |
| 1 | 5 | 0.07095 | 242 | 10 | 0.53823 | 0.98511 | 3.43106 | 0.47567 |
| 2 | 6 | 0.13148 | 121 | 10 | 0.52493 | 0.91239 | 4.40584 | 0.19543 |
| 3 | 7 | 0.15575 | 102 | 9 | 0.50009 | 0.80159 | 1.43484 | 0.98014 |
| 4 | 7 | 0.10178 | 245 | 9 | 0.50621 | 0.55022 | 2.84160 | 0.78872 |
| 5 | 6 | 0.15994 | 88 | 9 | 0.54667 | 0.93870 | 0.68681 | 0.82277 |
| 6 | 6 | 0.07483 | 221 | 7 | 0.50084 | 0.97352 | 3.24507 | 0.65469 |
| 7 | 7 | 0.08191 | 179 | 8 | 0.50132 | 0.99302 | 4.00168 | 0.63458 |
| 8 | 5 | 0.05154 | 256 | 10 | 0.50279 | 0.97827 | 1.93594 | 0.01850 |
| 9 | 8 | 0.05644 | 294 | 10 | 0.56394 | 0.84780 | 2.05943 | 0.27115 |
| 10 | 4 | 0.07670 | 281 | 10 | 0.53927 | 0.97848 | 4.82450 | 0.27244 |
| 11 | 6 | 0.13550 | 112 | 10 | 0.53406 | 0.53254 | 2.40913 | 0.53195 |
| 12 | 6 | 0.08587 | 203 | 10 | 0.63046 | 0.84871 | 4.82865 | 0.96621 |
| 13 | 7 | 0.09585 | 288 | 10 | 0.50229 | 0.99575 | 4.49999 | 0.55975 |
| 14 | 7 | 0.08191 | 245 | 10 | 0.54189 | 0.87572 | 3.95663 | 0.85872 |
| 15 | 7 | 0.07474 | 174 | 7 | 0.51174 | 0.96561 | 2.97146 | 0.92806 |
| 16 | 5 | 0.08019 | 212 | 9 | 0.50228 | 0.98434 | 4.45686 | 0.41716 |
| 17 | 8 | 0.07642 | 205 | 9 | 0.52041 | 0.84692 | 4.46048 | 0.41196 |
| 18 | 8 | 0.08315 | 218 | 10 | 0.56530 | 0.92783 | 4.49138 | 0.69385 |
| 19 | 7 | 0.09760 | 171 | 10 | 0.57999 | 0.74319 | 0.76715 | 0.72171 |
| 20 | 5 | 0.10593 | 142 | 9 | 0.52512 | 0.99411 | 0.89520 | 0.27131 |
| 21 | 8 | 0.07079 | 185 | 9 | 0.50469 | 0.87378 | 1.08559 | 0.25444 |
| 22 | 6 | 0.11366 | 183 | 9 | 0.57435 | 0.77739 | 3.16044 | 0.93374 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 23 | 6 | 0.11157 | 254 | 9 | 0.52017 | 0.97489 | 1.90816 | 0.79666 |
| 24 | 7 | 0.11255 | 188 | 8 | 0.50796 | 0.74881 | 1.66455 | 0.77696 |
| 25 | 7 | 0.18193 | 79 | 10 | 0.68663 | 0.99816 | 2.79139 | 0.92738 |
| 26 | 5 | 0.20795 | 137 | 10 | 0.60099 | 0.37442 | 4.72568 | 0.01013 |
| 27 | 5 | 0.08039 | 208 | 10 | 0.55245 | 0.85163 | 2.68594 | 0.57202 |
| 28 | 4 | 0.13824 | 232 | 9 | 0.53167 | 0.97794 | 4.99790 | 0.72989 |
| 29 | 5 | 0.11709 | 264 | 10 | 0.50079 | 0.65333 | 4.99177 | 0.01211 |
| 30 | 7 | 0.08384 | 183 | 9 | 0.54315 | 0.85012 | 2.95216 | 0.68107 |
| 31 | 7 | 0.07624 | 243 | 10 | 0.51390 | 0.68864 | 2.39622 | 0.79548 |
| 32 | 8 | 0.12670 | 219 | 9 | 0.51735 | 0.51438 | 4.86510 | 0.39015 |
| 33 | 7 | 0.17030 | 133 | 8 | 0.50620 | 0.74374 | 4.58307 | 0.02592 |
| 34 | 6 | 0.17322 | 101 | 7 | 0.54173 | 0.74255 | 4.24794 | 0.97291 |
| 35 | 4 | 0.09476 | 222 | 10 | 0.50025 | 0.77689 | 4.46467 | 0.83712 |
| 36 | 7 | 0.10917 | 175 | 9 | 0.52491 | 0.91604 | 2.16957 | 0.73717 |
| 37 | 7 | 0.08104 | 168 | 10 | 0.50263 | 0.50745 | 3.81626 | 0.69286 |
| 38 | 5 | 0.13034 | 245 | 10 | 0.50188 | 0.76513 | 2.11312 | 0.03408 |
| 39 | 8 | 0.08419 | 196 | 10 | 0.55446 | 0.85748 | 4.85152 | 0.63630 |
| 40 | 8 | 0.08592 | 205 | 10 | 0.52833 | 0.88829 | 2.58534 | 0.47814 |
| 41 | 6 | 0.16963 | 80 | 8 | 0.56274 | 0.94076 | 3.32882 | 0.66880 |
| 42 | 7 | 0.14335 | 123 | 8 | 0.56362 | 0.91686 | 4.92034 | 0.02893 |
| 43 | 8 | 0.12178 | 183 | 10 | 0.58719 | 0.86078 | 3.42019 | 0.41184 |
| 44 | 7 | 0.14413 | 185 | 10 | 0.52721 | 0.57415 | 2.30624 | 0.51660 |
| 45 | 6 | 0.08748 | 221 | 10 | 0.50936 | 0.70774 | 2.06636 | 0.28648 |
| 46 | 7 | 0.08946 | 176 | 10 | 0.55001 | 0.92642 | 3.51959 | 0.19652 |
| 47 | 7 | 0.09190 | 157 | 8 | 0.63144 | 0.74802 | 0.11220 | 0.61326 |
| 48 | 5 | 0.13140 | 200 | 10 | 0.51145 | 0.69994 | 3.88528 | 0.53884 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 49 | 5 | 0.12585 | 187 | 10 | 0.50120 | 0.93257 | 2.44567 | 0.98524 |
| 50 | 6 | 0.07366 | 250 | 9 | 0.53270 | 0.95103 | 1.97326 | 0.23232 |

**Table S4.** Hyperparameter optimization results of all 50 runs for Group 1 of ETEX-1 experiment.

| Run | Hyperparameters | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | *max_depth* | *learning_rate* | *n_estimators* | *min_child_weight* | *subsample* | *colsample_bytree* | *reg_lambda* | *gamma* |
| 1 | 3 | 0.07742 | 63 | 9 | 0.55953 | 0.44389 | 0.35824 | 0.81392 |
| 2 | 4 | 0.05011 | 142 | 3 | 0.70683 | 0.23027 | 4.89201 | 0.98193 |
| 3 | 3 | 0.06110 | 99 | 2 | 0.93364 | 0.37012 | 2.20274 | 0.76370 |
| 4 | 7 | 0.06117 | 64 | 10 | 0.55117 | 0.24878 | 2.34822 | 0.07471 |
| 5 | 3 | 0.07906 | 76 | 9 | 0.71631 | 0.38280 | 1.84573 | 0.22755 |
| 6 | 3 | 0.05067 | 116 | 4 | 0.70755 | 0.37613 | 1.50917 | 0.85795 |
| 7 | 3 | 0.06296 | 86 | 9 | 0.76834 | 0.34797 | 1.04674 | 0.44097 |
| 8 | 3 | 0.05342 | 106 | 5 | 0.53520 | 0.18199 | 4.18213 | 0.89476 |
| 9 | 3 | 0.07584 | 76 | 4 | 0.95540 | 0.53685 | 1.30937 | 0.49309 |
| 10 | 3 | 0.07213 | 84 | 5 | 0.98527 | 0.41048 | 2.61014 | 0.06896 |
| 11 | 3 | 0.05907 | 115 | 6 | 0.74495 | 0.37051 | 1.96059 | 0.27702 |
| 12 | 3 | 0.09471 | 76 | 3 | 0.91290 | 0.51041 | 0.38558 | 0.05232 |
| 13 | 3 | 0.07018 | 103 | 3 | 0.87242 | 0.35732 | 2.77176 | 0.16073 |
| 14 | 3 | 0.07072 | 95 | 10 | 0.98317 | 0.34966 | 4.67025 | 0.95006 |
| 15 | 3 | 0.08357 | 66 | 2 | 0.80913 | 0.37858 | 2.74202 | 0.05494 |
| 16 | 3 | 0.05001 | 121 | 3 | 0.52214 | 0.27101 | 4.30584 | 0.30632 |
| 17 | 3 | 0.09354 | 50 | 6 | 0.64736 | 0.62473 | 2.55863 | 0.35745 |
| 18 | 3 | 0.05486 | 134 | 8 | 0.66206 | 0.71278 | 0.80280 | 0.97413 |
| 19 | 3 | 0.07556 | 102 | 5 | 0.70927 | 0.34789 | 3.12167 | 0.99997 |
| 20 | 3 | 0.07479 | 52 | 9 | 0.79240 | 0.56720 | 1.02323 | 0.32951 |
| 21 | 3 | 0.05518 | 78 | 10 | 0.66309 | 0.99871 | 3.14571 | 0.84078 |
| 22 | 3 | 0.05139 | 111 | 6 | 0.73839 | 0.42603 | 2.49218 | 0.87318 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 23 | 3 | 0.10952 | 50 | 5 | 0.97390 | 0.22350 | 0.88047 | 0.35097 |
| 24 | 3 | 0.06860 | 93 | 8 | 0.66447 | 0.33031 | 3.95098 | 0.67626 |
| 25 | 3 | 0.08670 | 54 | 5 | 0.79857 | 0.39303 | 3.19098 | 0.54197 |
| 26 | 3 | 0.08125 | 67 | 8 | 0.94963 | 0.33151 | 4.40350 | 0.06507 |
| 27 | 3 | 0.08396 | 50 | 2 | 0.93428 | 0.37792 | 3.77359 | 0.13881 |
| 28 | 3 | 0.05418 | 95 | 9 | 0.99598 | 0.25227 | 1.60204 | 0.38791 |
| 29 | 3 | 0.06935 | 89 | 8 | 0.90482 | 0.35876 | 4.15848 | 0.85423 |
| 30 | 3 | 0.06319 | 76 | 2 | 0.91583 | 0.43665 | 3.35600 | 0.88327 |
| 31 | 3 | 0.09009 | 50 | 4 | 0.94183 | 0.48645 | 4.03998 | 0.17582 |
| 32 | 3 | 0.06213 | 148 | 8 | 0.69235 | 0.34063 | 1.38470 | 0.71691 |
| 33 | 3 | 0.05735 | 74 | 10 | 0.83704 | 0.36175 | 2.28311 | 0.93893 |
| 34 | 3 | 0.06437 | 72 | 3 | 0.98748 | 0.31363 | 1.63480 | 0.22685 |
| 35 | 3 | 0.06022 | 60 | 6 | 0.66204 | 0.69317 | 1.21692 | 0.30900 |
| 36 | 3 | 0.09555 | 53 | 5 | 0.80980 | 0.46487 | 1.90000 | 0.60232 |
| 37 | 3 | 0.08434 | 50 | 8 | 0.52774 | 0.26641 | 0.48391 | 0.31574 |
| 38 | 3 | 0.07105 | 51 | 5 | 0.96131 | 0.63725 | 2.01205 | 0.60509 |
| 39 | 4 | 0.05005 | 62 | 10 | 0.77964 | 0.22026 | 3.55884 | 0.74839 |
| 40 | 4 | 0.07553 | 74 | 2 | 0.99323 | 0.36292 | 2.61782 | 0.17595 |
| 41 | 3 | 0.05239 | 77 | 7 | 0.50028 | 0.95751 | 2.41469 | 0.72211 |
| 42 | 3 | 0.08421 | 51 | 5 | 0.99977 | 0.45864 | 2.15063 | 0.54258 |
| 43 | 3 | 0.05414 | 82 | 8 | 0.62226 | 0.76122 | 2.83002 | 0.53414 |
| 44 | 3 | 0.05259 | 111 | 4 | 0.93432 | 0.32197 | 2.04760 | 0.44156 |
| 45 | 3 | 0.09454 | 60 | 4 | 0.72047 | 0.23879 | 4.65624 | 0.75740 |
| 46 | 3 | 0.05013 | 93 | 4 | 0.58250 | 0.40493 | 2.11383 | 0.47864 |
| 47 | 3 | 0.11330 | 50 | 2 | 0.71413 | 0.69524 | 3.50503 | 0.16269 |
| 48 | 3 | 0.08314 | 50 | 10 | 0.96691 | 0.50529 | 2.97909 | 0.95771 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 49 | 6 | 0.05978 | 107 | 7 | 0.63666 | 0.20488 | 0.61715 | 0.79254 |
| 50 | 3 | 0.07770 | 71 | 3 | 0.98943 | 0.58108 | 1.17867 | 0.22360 |

**Table S5.** Hyperparameter optimization results of all 50 runs for Group 2 of ETEX-1 experiment.

| Run | Hyperparameters | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | *max_depth* | *learning_rate* | *n_estimators* | *min_child_weight* | *subsample* | *colsample_bytree* | *reg_lambda* | *gamma* |
| 1 | 8 | 0.17252 | 127 | 7 | 0.58651 | 0.60647 | 4.02984 | 0.60724 |
| 2 | 8 | 0.22988 | 68 | 9 | 0.54811 | 0.91321 | 2.29180 | 0.12852 |
| 3 | 8 | 0.10002 | 213 | 10 | 0.75238 | 0.64053 | 4.99653 | 0.10778 |
| 4 | 4 | 0.12366 | 147 | 5 | 0.87463 | 0.83119 | 0.63085 | 0.26995 |
| 5 | 5 | 0.23441 | 297 | 7 | 0.97266 | 0.36675 | 0.34489 | 0.99034 |
| 6 | 4 | 0.20533 | 155 | 4 | 0.63974 | 0.56526 | 2.63317 | 0.38177 |
| 7 | 5 | 0.10641 | 151 | 3 | 0.76094 | 0.79390 | 0.98025 | 0.96160 |
| 8 | 4 | 0.17290 | 222 | 4 | 0.55589 | 0.76284 | 1.62191 | 0.47379 |
| 9 | 4 | 0.05855 | 160 | 3 | 0.88818 | 0.79781 | 4.98019 | 0.84983 |
| 10 | 4 | 0.11741 | 184 | 5 | 0.79714 | 0.83203 | 4.43324 | 0.62777 |
| 11 | 4 | 0.16266 | 136 | 2 | 0.75690 | 0.93818 | 3.30854 | 0.55481 |
| 12 | 5 | 0.23134 | 144 | 2 | 0.58373 | 0.60423 | 2.76711 | 0.16986 |
| 13 | 8 | 0.27891 | 193 | 7 | 0.64977 | 0.89059 | 3.88204 | 0.23152 |
| 14 | 7 | 0.18603 | 245 | 8 | 0.77290 | 0.78709 | 3.45149 | 0.01806 |
| 15 | 5 | 0.16915 | 268 | 10 | 0.62385 | 0.49651 | 2.30355 | 0.27120 |
| 16 | 4 | 0.20217 | 64 | 2 | 0.92784 | 0.78470 | 0.94699 | 0.93657 |
| 17 | 5 | 0.10871 | 181 | 9 | 0.70489 | 0.84917 | 4.43678 | 0.07228 |
| 18 | 5 | 0.07394 | 297 | 8 | 0.87839 | 0.62200 | 3.24008 | 0.11160 |
| 19 | 6 | 0.20293 | 216 | 9 | 0.66381 | 0.89210 | 4.08151 | 0.60613 |
| 20 | 7 | 0.20570 | 158 | 4 | 0.50653 | 0.86393 | 3.36667 | 0.79227 |
| 21 | 5 | 0.20083 | 88 | 7 | 0.57460 | 0.62410 | 1.26707 | 0.17321 |
| 22 | 4 | 0.27072 | 50 | 4 | 0.85604 | 0.86560 | 0.16264 | 0.44052 |

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|---|---|---|---|---|---|---|---|---|
| 23 | 8 | 0.15380 | 86 | 5 | 0.67811 | 0.74505 | 4.54334 | 0.93377 |
| 24 | 4 | 0.16205 | 183 | 6 | 0.59364 | 0.93969 | 1.05664 | 0.40669 |
| 25 | 6 | 0.14171 | 288 | 6 | 0.75389 | 0.85527 | 4.65363 | 0.50557 |
| 26 | 7 | 0.21287 | 253 | 9 | 0.59311 | 0.65113 | 2.79234 | 0.83703 |
| 27 | 4 | 0.15371 | 247 | 5 | 0.77890 | 0.52357 | 4.81584 | 0.67752 |
| 28 | 5 | 0.11665 | 135 | 5 | 0.79729 | 0.86017 | 4.26743 | 0.12912 |
| 29 | 4 | 0.08378 | 192 | 4 | 0.52749 | 0.79980 | 2.64816 | 0.57092 |
| 30 | 6 | 0.13030 | 210 | 3 | 0.50209 | 0.61548 | 3.80894 | 0.64347 |
| 31 | 7 | 0.24148 | 173 | 10 | 0.64711 | 0.79358 | 2.66441 | 0.23023 |
| 32 | 6 | 0.09301 | 204 | 8 | 0.69879 | 0.97301 | 4.67770 | 0.36945 |
| 33 | 5 | 0.12318 | 283 | 6 | 0.93580 | 0.70267 | 2.23369 | 0.17565 |
| 34 | 4 | 0.23289 | 227 | 7 | 0.60924 | 0.76662 | 2.97809 | 0.22066 |
| 35 | 6 | 0.21219 | 162 | 3 | 0.54969 | 0.50796 | 4.01790 | 0.10632 |
| 36 | 5 | 0.10657 | 148 | 3 | 0.77407 | 0.84022 | 4.19435 | 0.53237 |
| 37 | 4 | 0.14220 | 169 | 4 | 0.69411 | 0.90516 | 2.46148 | 0.83182 |
| 38 | 6 | 0.11224 | 239 | 4 | 0.64335 | 0.91879 | 1.53421 | 0.43750 |
| 39 | 5 | 0.08990 | 98 | 5 | 0.83843 | 0.99546 | 3.80815 | 0.86071 |
| 40 | 6 | 0.19006 | 130 | 4 | 0.95749 | 0.88483 | 3.68950 | 0.17261 |
| 41 | 5 | 0.21434 | 93 | 6 | 0.80593 | 0.97025 | 2.23769 | 0.40479 |
| 42 | 7 | 0.07619 | 234 | 6 | 0.62146 | 0.77954 | 4.62217 | 0.85628 |
| 43 | 4 | 0.17377 | 273 | 6 | 0.85218 | 0.79578 | 3.43808 | 0.62076 |
| 44 | 4 | 0.18522 | 135 | 4 | 0.82615 | 0.63563 | 4.24215 | 0.56409 |
| 45 | 4 | 0.14993 | 152 | 8 | 0.60441 | 0.80580 | 2.50467 | 0.09351 |
| 46 | 5 | 0.15229 | 164 | 7 | 0.94667 | 0.83661 | 3.59476 | 0.15891 |
| 47 | 5 | 0.15393 | 116 | 9 | 0.90651 | 0.85377 | 4.60433 | 0.89894 |
| 48 | 5 | 0.22272 | 290 | 9 | 0.86799 | 0.85502 | 4.52637 | 0.79836 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 49 | 5 | 0.11275 | 91 | 4 | 0.72730 | 0.75528 | 3.72672 | 0.17298 |
| 50 | 4 | 0.13702 | 299 | 7 | 0.95702 | 0.91622 | 2.93120 | 0.22371 |

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